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A quadrature-based conditional moment closure for mixing-sensitive reactions

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Abstract

A novel algorithm consisting of a quadrature-based semi-analytical solution to the conditional moment closure (CMC) is developed for mixing-sensitive reactions in turbulent flows. When applying the proposed algorithm, the additional grid in mixture-fraction phase space used in CMC codes is eliminated, and at most ten quadrature nodes are needed to model mixing-sensitive turbulent reacting flows. In this work, the mixture-fraction probability density function (PDF) is assumed to be a β-PDF, which is the weight function for the Gauss-Jacobi quadrature rule. The conditional moments of reacting species are determined from unconditional moments that are first order with respect to the species and higher order with respect to mixture fraction. Here, the focus is on the efficient treatment of the molecular-mixing step by using a semi-analytical solution in the form of a Jacobi polynomial expansion. The application of the algorithm is demonstrated considering mixing-sensitive competitive-consecutive and parallel reactions in a statistically homogeneous chemical reactor.

Keywords

mixing-sensitive reactions, conditional moment closure, quadrature based-solution algorithm, turbulent mixing

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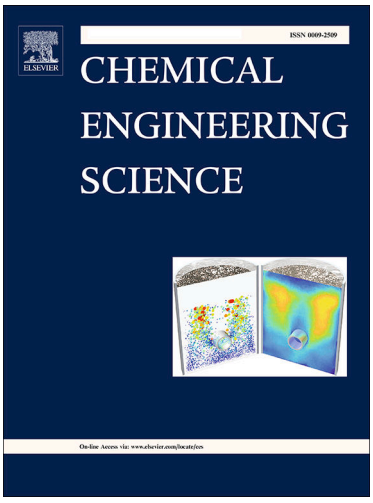

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A quadrature-based conditional moment closure for mixing-sensitive reactions

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ABSTRACT

A novel algorithm consisting of a quadrature-based semi-analytical solution to the conditional moment closure (CMC) is developed for mixing-sensitive reactions in turbulent flows. When applying the proposed algorithm, the additional grid in mixture-fraction phase space used in CMC codes is eliminated, and at most ten quadrature nodes are needed to model mixing-sensitive turbulent reacting flows. In this work, the mixture-fraction probability density function (PDF) is assumed to be a β-PDF, which is the weight function for the Gauss-Jacobi quadrature rule. The conditional moments of reacting species are determined from unconditional moments that are first order with respect to the species and higher order with respect to mixture fraction. Here, the focus is on the efficient treatment of the molecular-mixing step by using a semi-analytical solution in the form of a Jacobi polynomial expansion. The application of the algorithm is demonstrated considering mixing-sensitive competitive-consecutive and parallel reactions in a statistically homogeneous chemical reactor.

Keywords: mixing-sensitive reactions; conditional moment closure; quadrature based-solution algorithm; turbulent mixing

1. Introduction

For turbulent mixing with finite-rate chemistry, important parameters such as reaction yield and mean species concentrations are difficult to predict accurately because, in addition to a range of reaction time scales, the spatial inhomogeneity, and unresolved concentration fluctuations prevent the straightforward solution of the species conservation equations (Bourne 2003). Moreover, the complexity of the problem becomes more acute when the chemical species have different molecular diffusivities, especially in high Schmidt number turbulent flows (Fox 2003).

In order to overcome these challenges, several different modeling approaches have been proposed in the literature such as (i) the transported probability density function (PDF) approach

(Kuron et al. 2017, Madadi-Kandjani et al. 2017, Raman et al. 2005), (ii) the presumed PDF approach (Bray et al. 2006, Libby and Williams, 1980), (iii) the conditional moment closure (Smith et al. 1992, Kronenburg and Mastorakos 2011, Fairweather and Wooley 2003, Klimenko and Bilger 1999), and (iv) moment closures. Among them, the first three utilize the composition PDF to describe the turbulent statistics and have gained tremendous attention because the Reynolds-average chemical source term in these methods does not require any further modeling (Fox 2003). On the other hand, the molecular-mixing term requires modeling, and this issue has been studied by many authors (Villermaux and Devillon 1972, Janicka et al. 1979, Nooren et al. 1997, Subramaniam and Pope 1998, Fox, 1992, Fox 1994, Fox and Yeung 2003, Pope 1991), especially with transported PDF methods (Klimenko and Pope 2003, Madadi-Kandjani et al. 2017). However, in the context of computational fluid dynamics (CFD), the use of the latter method is affordable only if there are a few chemical species. For this reason, presumed PDF and CMC methods have gained more attention since the use of many species is computationally tractable due to the form of these closures. Furthermore, CMC has the advantage over presumed PDF methods since only the form of mixture-fraction (or conserved scalar) PDF must be assumed, while, in the presumed PDF approach, the form of the joint PDF must be supplied (which must be consistent with the mixture-fraction PDF).

The CMC method employed in this work was proposed independently by Bilger (1993) and Klimenko (1995) and is based on the idea that, if there is no correlation between the conditional scalar dissipation rate (CSDR) and the reaction-progress variable, the fluctuations around the conditional mean are negligible when conditioned on the mixture fraction. This idea allows one to estimate the conditional chemical source term in terms of the first-order conditional moments of the reactants by neglecting the conditional covariances. Then, it is straightforward to calculate the unconditional chemical source terms using the assumed form of the mixture-fraction PDF. However, when implemented in a CFD code, this approach requires an additional grid for the mixture-fraction phase space, i.e., due to conditioning, the dimensionality of the problem is increased. When simulating plant-scale reactors with non-trivial geometry using CFD, this additional dimension greatly increases the computational cost. Here, our goal is to reduce this cost by tracking a small set of composition moments, which are used to find the conditional moments via Gaussian quadrature.

One of the first study dealing with the application of quadrature-based moment methods (QBMM) to CMC (Smith et al. 2007) focused on the quadrature approximation to the conditional reaction-source term. Earlier, Fox (2003) proposed an extension of the multi-environment PDF (ME-PDF) method, called multi-environment conditional PDF model (MEC-PDF) by using conditional scalar means that was subsequently solved by the direct quadrature method of moments (Fox and Raman 2004). As with the ME-PDF model, when the chemistry is sensitive to the shape of the joint composition PDF, the number of environments must be chosen accordingly. However, when more than two environments are needed to describe a turbulent reacting flow, the molecular-mixing model must be carefully chosen to describe the higher-order moments for the composition variables (Madadi-Kandjani et al. 2017).

To improve the representation of molecular mixing in multi-environment models with more than two environments, in this work, we start from the CMC governing equation and propose a semi-analytical solution (SA-CMC) for molecular mixing written in terms of a Jacobi polynomial expansion. This is done by first introducing a deviation variable for each composition variable, and semi-analytically solving its time variation in mixture-fraction space. The Jacobi polynomial expansion is paired with a β-PDF for the mixture fraction, and hence a Gaussian quadrature rule is used to evaluate integrals (e.g., first-order composition moments) with respect to the β-PDF. From a computational standpoint, this mathematical representation is convenient because the values of the first-order conditional moments for the chemical species are only needed at the quadrature nodes (and not at every point in mixture-fraction phase space).

In this work, the application of the proposed quadrature-based algorithm is illustrated for mixing-sensitive, competitive-consecutive and parallel reactions in a statistically homogeneous chemical reactor. Nonetheless, its extension to CFD simulations of inhomogeneous systems is straightforward and involves solving the transport equations for the unconditional moments, followed by the application of the quadrature-based algorithm for each cell in the mesh. With this in mind, the rest of the paper is organized as follows. After an introductory description of the CMC method, Secs. 3 and 4 present the quadrature-based algorithm. Subsequently, Secs. 5 and 6 are devoted to the test cases and the simulation results, respectively. In Sec. 7, relevant conclusions are drawn, and future research directions are proposed.

2. Conditional Moment Models for Turbulent Reacting Flows

In this section, we provide a brief overview of models for turbulent reacting flows based on first-order concentration moments conditioned on the mixture fraction and describe how these are related to Gaussian quadrature through CQMOM. Because each chemical species is treated in the same manner in the molecular-mixing step, for clarity, we will develop the quadrature-based algorithm for a single conditional scalar mean. However, in the chemical source term, all the chemical species are coupled through the reactions, and hence their composition variables must be updated using a coupled ordinary differential equation (ODE) solver designed for chemical kinetics.

2.1 Conditional Moment Closure

In CMC, a five-dimensional transport equation is solved for the conditional scalar mean, $Q(\zeta;x,t)$ that is defined as

$$Q(\zeta;x,t)=\left\langle \phi(x,t)\middle|\zeta\right\rangle=\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\psi f_{\phi|\xi}\left(\psi\middle|\zeta;x,t\right)d\psi \tag{2.5}$$

where x is the 3-D spatial variable, ζ is the 1-D sample space variable for the mixture fraction, and t is time. $f_{\phi|\xi}\left(\psi\middle|\zeta;x,t\right)$ is the conditional PDF of the composition variable $\phi(x,t)$ given the mixture fraction, while ψ is the corresponding sample space variable. For a spatially homogeneous flow wherein the turbulence statistics do not depend on the physical location, the CMC transport equation for $Q(\zeta;t)$ can be written in terms of the variable $X(\zeta,t)=Q(\zeta;t)f_{\xi}(\zeta,t)$

$$\frac{\partial X}{\partial t}=\frac{\partial}{\partial\zeta}\left[Yf_{\xi}\frac{\partial Q}{\partial\zeta}-Q\frac{\partial\left(Yf_{\xi}\right)}{\partial\zeta}\right]+S(Q)f_{\xi} \tag{2.6}$$

where $f_{\xi}(\zeta;t)$ is the mixture-fraction PDF with sample-space variable ζ, and $Y(\zeta,t)$ is the conditional scalar dissipation rate (CSDR). The first term on the right-hand side (RHS) of Eq. (2.2) represents molecular mixing in mixture-fraction phase space, and its form is the same for each chemical species. The second term on the RHS of Eq. (2.2) represents changes due to chemical reactions, and it is closed given the vector *Q* representing all of the chemical species. As mentioned

above, the main focus of this work is the solution of the molecular-mixing term in Eq. (2.2). Thus, for clarity, from now on in this section, we take Q to be a scalar.

For statistically homogeneous flows (i.e. $f_\xi = f_\xi(\zeta,t)$) the mixture-fraction PDF evolves according to

$$\frac{\partial f_\xi}{\partial t} = -\frac{\partial^2 Y f_\xi}{\partial \zeta^2} \tag{2.7}$$

In this work, the mixture-fraction PDF is assumed to follow a β-PDF:

$$f_\xi = \frac{\zeta^{a-1}(1-\zeta)^{b-1}}{\beta(a,b)} \tag{2.8}$$

where $a = \langle\xi\rangle/\sigma$ and $b = 1-\langle\xi\rangle/\sigma$ with $\sigma = \left(\langle\xi^2\rangle - \langle\xi\rangle^2\right)\Big/\left(\langle\xi\rangle - \langle\xi^2\rangle\right)$. In a non-premixed system, the initial conditions are such that a and b are null, and thus at time zero σ is infinite. For long times, a and b approach infinity, so that σ goes to zero for a perfectly mixed system. Note that during molecular mixing the mean mixture fraction $\langle\xi\rangle$ is constant so that σ changes due to the second-order moment $\langle\xi^2\rangle$. As is usually done in presumed PDF methods, in CFD applications transport equations will be solved for $\langle\xi\rangle$ and $\langle\xi^2\rangle$, so that their values are known in each grid cell.

As is shown by Tsai and Fox (1995), in CMC the form of the CSDR cannot be chosen independently from the mixture-fraction PDF. This is because if the CSDR does not correspond to the presumed mixture-fraction PDF, then the unconditional moment computed from Q will not be constant during molecular mixing as it should be due to the conservation of chemical species. For a β-PDF, the CSDR used in the Fokker-Plank model (Fox 2003) has the form

$$Y(\zeta,t) = \frac{\varepsilon_\xi}{2}\left(\frac{\zeta-\zeta^2}{\langle\xi\rangle - \langle\xi^2\rangle}\right) = \frac{1}{2}\frac{\sigma}{\tau_\xi}\left(\zeta-\zeta^2\right) \tag{2.9}$$

where ε_ξ is the scalar dissipation rate (SDR) and τ_ξ is the molecular mixing timescale. In general, the mixture-fraction moments are defined by $\left\langle \xi^k \right\rangle = \int_0^1 \zeta^k f_\xi(\zeta;x,t)\,d\zeta$. From this definition, it is straightforward to show starting from Eq. (2.3) and using Eq. (2.5) that

$$\frac{\partial\langle\xi\rangle}{\partial t} = 0, \qquad \frac{\partial\langle\xi^2\rangle}{\partial t} = -2\langle Y\rangle = -\varepsilon_\xi = \frac{\langle\xi\rangle^2 - \langle\xi^2\rangle}{\tau_\xi} \tag{2.10}$$

which confirms that the mean mixture fraction is constant, and its second moment is a decreasing function of the SDR. The molecular-mixing timescale depends on the turbulence timescales. In this work, we set $\tau_\xi = 1\text{s}$ so that all chemical reaction rates are expressed relative to this value. In a CFD simulation of a turbulent reacting flow, the SDR in each grid cell is determined from the local turbulence integral timescale (Liu and Fox 2006), thereby coupling the turbulent flow field to the chemistry.

CMC gives accurate results if the conditional fluctuations around the conditional scalar mean are negligible. This assumption allows to approximate the conditional chemical source term through $\left\langle S(\phi;x,t)\middle|\xi\right\rangle \approx S\left(\left\langle\phi\middle|\xi=\zeta;x,t\right\rangle\right) \equiv S\left(Q(\zeta;x,t)\right)$, and the Reynolds-averaged chemical source term can then be found from

$$\left\langle S(\phi)\right\rangle(x,t) = \int_0^1 S\left(Q(\zeta;x,t)\right) f_\xi(\zeta;x,t)\,d\zeta \tag{2.11}$$

where Q is the vector of conditional moments for all chemical species in the flow. Additionally, the unconditional scalar mean for each chemical species can be found from

$$\langle\phi\rangle(x,t) = \int_0^1 Q(\zeta;x,t)\, f_\xi(\zeta;x,t)\,d\zeta \tag{2.12}$$

For the β-PDF, the integral over the mixture-fraction phase space can be computed using a Gauss-Jacobi quadrature rule (Gautschi 2004), which is uniquely determined from the known values of a and b.

2.2 Definition of moments and the quadrature-based approximation

The composition moments in their generic form are defined as

$$\left\langle g\left(\phi\right)\right\rangle = \int_{\Omega_\psi} g\left(\psi\right) f_\phi\left(\psi;x,t\right) d\psi \tag{2.13}$$

such that the mean composition given in Eq. (2.12) can be found with $g\left(\psi\right)=\psi$. Meanwhile, the joint moments $\left\langle \xi^k \phi \right\rangle$ are defined similarly with $g\left(\psi\right)=\zeta^k\psi$ and $f_{\phi,\xi} = f_{\phi|\xi} f_\xi$:

$$\left\langle \xi^k \phi \right\rangle = \int_0^1 \int_{-\infty}^{\infty} \zeta^k \psi f_{\phi|\xi}\left(\psi|\zeta\right) f_\xi\left(\zeta;x,t\right) d\psi d\zeta \tag{2.14}$$

where $f_{\phi|\xi}\left(\psi|\zeta\right)$ is the conditional composition PDF conditioned on the mixture fraction. For first-order CMC, this term is approximated via Dirac delta functions as

$$f_{\phi|\xi}\left(\psi|\zeta;x,t\right) \cong \delta\left(\psi - \left\langle \phi|\zeta \right\rangle\right) \equiv \delta\left(\psi - Q\left(\zeta;x,t\right)\right) \tag{2.15}$$

Thus, in terms of $X\left(\zeta,t\right)$, the species moments can be written as

$$\left\langle \xi^k Q \right\rangle = \int_0^1 \zeta^k X d\zeta \tag{2.16}$$

Using Eq. (2.11) in Eq. (2.10), the joint mixture-fraction, chemical-species moments $\left\langle \xi^k \phi \right\rangle$ are equivalent to the moments $\left\langle \xi^k Q \right\rangle$. The latter are approximated via the Gauss-Jacobi quadrature rule:

$$\left\langle \xi^k Q \right\rangle = \sum_{i=1}^{N} w_i \xi_i^k Q_i \tag{2.17}$$

where the w_i are the weights and ξ_i are the corresponding abscissas, which are known given the mean and variance of the β-PDF for the mixture fraction.

Using CQMOM, Eq. (2.17) yields a linear system for $Q_i = Q\left(\xi_i,t\right)$, i.e., the conditional chemical species concentration evaluated at the mixture-fraction abscissas, where the joint moments $\left\langle \xi^k Q \right\rangle$ are known:

$$\begin{bmatrix} 1 & 1 & \cdots & 1 \\ \xi_1 & \xi_2 & \cdots & \xi_N \\ \vdots & \vdots & \ddots & \vdots \\ \xi_1^{N-1} & \xi_2^{N-1} & \cdots & \xi_N^{N-1} \end{bmatrix} \begin{bmatrix} w_1 & & & \\ & w_2 & & \\ & & \ddots & \\ & & & w_N \end{bmatrix} \begin{bmatrix} Q_1 \\ Q_2 \\ \vdots \\ Q_N \end{bmatrix} = \begin{bmatrix} \left\langle \phi \right\rangle \\ \left\langle \xi Q \right\rangle \\ \vdots \\ \left\langle \xi^{N-1} Q \right\rangle \end{bmatrix} \tag{2.18}$$

Readers familiar with CQMOM (Cheng and Fox 2010, (Yuan and Fox 2011) will recognize the column vector on the LHS of Eq. (2.18) as the conditional moments, which can be found by solving Eq. (2.18) with an appropriate linear solver. This linear system has a unique solution because the abscissas are distinct, and all the weights are positive for a Gaussian quadrature. However, for large N (e.g., $N>5$), the solution of Eq. (2.18) is prone to numerical errors when the leading matrix on the LHS has high condition number, requiring error-minimizing linear equation solvers. Furthermore, this numerical error increases with the number of nodes; thus, caution must be taken to prevent non-physical values of Q_i for large N.

3. Semi-analytical Solution for CMC

Our solution algorithm, described in Sec. 4, is based on operator splitting where the molecular mixing and chemical reactions are treated separately for each time interval. The molecular mixing part of the CMC governing equation is

$$\frac{\partial X}{\partial t}=\frac{\partial}{\partial \zeta}\left[Zf_{\xi}\frac{\partial Q}{\partial \zeta}-Q\frac{\partial\left(Zf_{\xi}\right)}{\partial \zeta}\right] \tag{3.20}$$

The semi-analytical (SA) solution is written in terms of the deviation variable $q(\zeta,t)$, which is defined as

$$Q(t,\zeta)=q(t,\zeta)+\left[Q(1)-Q(0)\right]\zeta+Q(0) \tag{3.21}$$

where $Q(1)$ and $Q(0)$ are the known boundary conditions for the chemical species. Thus, q represents the deviation of conditional scalar mean from pure mixing and is exactly zero on the boundaries, i.e., $q(t,0)=q(t,1)=0$. For that reason, it is convenient to solve for the deviation variable when operator splitting is used because it generates a homogenous differential equation. Moreover, its unconditional mean is conserved (i.e., $\langle q\rangle=$constant) during the molecular-mixing step. In any case, knowledge of $q(t,\zeta)$ is sufficient to find the conditional mean of the corresponding chemical species Q by using Eq. (3.2).

In this study, the mixture-fraction PDF is assumed to be a β-PDF, and the deviation variable is modeled with the following Jacobi differential equation (Zwillinger 1997):

$$\frac{\partial q}{\partial \tau} = \frac{1}{2}\left(1-x^2\right)\frac{\partial^2 q}{\partial x^2} + \kappa_1(x)\frac{\partial q}{\partial x} + \kappa_0(x)q \tag{3.22}$$

where $x = 2\zeta - 1$ (hence $x \in [-1,1]$), the rescaled time variable is

$$d\tau = \frac{\sigma}{\tau_\xi}dt \tag{3.23}$$

and the terms involving $\kappa(x)$ are

$$\kappa_1(x) = \frac{\langle\xi\rangle - 1}{\sigma} - \left(\frac{1}{2\sigma} - 1\right)x, \qquad \kappa_0(x) = 1 + \frac{x}{\left(1-x^2\right)}\left[\frac{2\langle\xi\rangle - 1}{\sigma} - \left(\frac{1}{\sigma} - 2\right)x\right] \tag{3.24}$$

The first term on the RHS of Eq. (3.3) is the standard CMC model for molecular mixing that makes use of the CSDR in Eq. (2.5), and the two terms involving $\kappa(x)$ are needed to ensure that $\langle q\rangle(t) = \langle q\rangle$ is constant for all times.

As shown in Madadi-Kandjani et al. (2017), the β-PDF for the mixture fraction is not exact at short times and, hence, these additional terms correct for this difference while enforcing the constraint that the mean value is invariant with time. Mathematically, it is well known that the analytical solution to Eq. (3.3) can be written in terms of an infinite sum of Jacobi polynomials, which are orthogonal with respect to a β-PDF. At short times, a large number of terms are required in the sum in order to capture the non-premixed initial conditions. However, since in most applications *q* is zero initially (i.e., it is nonzero due to the chemistry), it should be possible to use a relatively small number of terms without loss of accuracy.

Here, we seek an approximate solution to Eq. (3.3), valid over a short time interval, given an initial condition for the deviation variable. This SA-CMC solution will be valid when σ does not change too much over the time interval of interest. Under these conditions, we can assume that the β-PDF parameters *a* and *b* are known (i.e., the mean and variance of the mixture fraction are known) and vary slowly with time compared to *q*. By defining an n^{th} order polynomial in terms of the n^{th} order Jacobi polynomial as

$$\varphi_n(\zeta) := P_n^{(b,a)}(2\zeta - 1) \tag{3.25}$$

the SA-CMC approximation to the solution of Eq. (3.22) is

$$q(t,\zeta) = \langle q \rangle \frac{\zeta(1-\zeta)}{\langle \xi \rangle - \langle \xi^2 \rangle} \left[1 + \sum_{n=1}^{N} A_n e^{-\frac{1}{2} n(n+a+b+1)\tau} \varphi_n(\zeta) \right] \tag{3.26}$$

Note that this solution is exact if N is infinite, so using a finite value of N leads to a truncation error that is largest for short times (in the absence of chemical reactions).

Using the orthogonality of the Jacobi polynomials, the expansion coefficients A_n can be calculated from the following set of linear equations:

$$\begin{aligned}
\left\langle \xi^2 (1-\xi) \varphi_1(\xi) \right\rangle A_1 &= \frac{\langle \xi q \rangle}{\langle q \rangle} \langle \xi - \xi^2 \rangle - \langle \xi^2 - \xi^3 \rangle \\
\sum_{n=1}^{2} \left\langle \xi^3 (1-\xi) \varphi_n(\xi) \right\rangle A_n &= \frac{\langle \xi^2 q \rangle}{\langle q \rangle} \langle \xi - \xi^2 \rangle - \langle \xi^3 - \xi^4 \rangle \\
&\vdots \\
\sum_{n=1}^{N} \left\langle \xi^{N+1} (1-\xi) \varphi_n(\xi) \right\rangle A_n &= \frac{\langle \xi^N q \rangle}{\langle q \rangle} \langle \xi - \xi^2 \rangle - \langle \xi^{N+1} - \xi^{N+2} \rangle
\end{aligned} \tag{3.27}$$

The LHS coefficients can be calculated analytically in terms of the beta function:

$$\left\langle \xi^{j+1} (1-\xi) P_n^{(b,a)}(2\xi - 1) \right\rangle = \begin{cases} \dfrac{B(n+b+1, j+a+1)}{B(a,b)} \dbinom{j}{n} & j \geq n \\ 0 & j < n \end{cases} \tag{3.28}$$

The RHS of Eq. (3.8) are known from the initial conditions (i.e., the joint moments at the beginning of the time step) and the moments of the mixture-fraction PDF. The coefficient matrix on the LHS of Eq. (3.8) is lower triangular and the diagonal elements are positive due to Eq. (3.9). Thus, it is easily inverted using forward substitution. However, the condition number of the matrix increases rapidly with N, so care must be taken to properly scale the system when solving for A_i. Likewise, when a and b are large, the ratio of the beta functions in Eq. (3.9) must be computed with care to avoid round-off errors.

In the solution algorithm in Sec. 4, Eq. (3.7) is used to advance q over a time step Δt. The exponential term in Eq. (3.26) is thus written in terms of a scaled time step $\Delta\tau$, defined by

$$\Delta\tau(t) = \int_{t}^{t+\Delta t} \frac{\sigma}{\tau_\xi} ds \tag{3.29}$$

Due to the orthogonality properties of the Jacobi polynomials, the mean of the deviation variable is conserved, i.e., $\langle q \rangle(t) = \langle q \rangle$. At the beginning of the time step, the deviation moments $\langle \xi^k q \rangle$ on the RHS of Eq. (3.27) are defined as

$$\langle \xi^k q \rangle = \langle \xi^k Q \rangle - \left[Q(1) - Q(0) \right] \langle \xi^{k+1} \rangle - Q(0) \langle \xi^k \rangle \tag{3.30}$$

where the first term on the RHS is approximated from the previous time step using the Gauss-Jacobi quadrature rule whose weight function is the β-PDF (Gautschi 2004). Finally, it is noteworthy that the updated value of q from Eq. (3.7) is only needed at the mixture-fraction abscissas ξ_i, i.e., $q(\Delta t, \xi_i)$. This fact greatly reduces the truncation errors associated with using a finite value for N in the SA-CMC approximation. In fact, the CQMOM linear system in Eq. (2.14) is equivalent to Eq. (3.8), and thus either system can be used to find the conditional abscissas $q_i = q(0, \xi_i)$ from the joint moments. However, for small N, the CQMOM system is more convenient to work with because the Jacobi polynomials do not need to be evaluated at the mixture-fraction abscissas.

4. Quadrature-Based Solution Algorithm

As noted in Sec. 3, operator splitting is the preferred method in CMC applications for treating spatial transport, molecular mixing, and chemical reactions over a time step Δt. Spatial transport is done with a CFD code to solve scalar transport equations for the unconditional moments of the mixture fraction and the chemical species. Here, only the molecular mixing and chemistry steps are described as the solution of moment transport equations can be done with any moment-preserving CFD solver such as OpenQBMM (Passalacqua et al., 2019). Note that unlike in the LES/CMC code where a coarser spatial grid is used to solve for the conditional moments due to the high computational cost of using a grid in mixture-fraction space (Zhang and Mastorakos 2016), with SA-CMC the same spatial grid can be used for all flow quantities. This difference will

be especially important for liquid-phase mixing where the large Schmidt number implies that all of the molecular mixing occurs at the sub-grid scales (Fox 2003).

The quadrature rule for the mixture-fraction weights and abscissas employs the routines developed by Gautschi (2004). For fixed values of *a* and *b*, one of three variants can be used: (1) Gauss-Jacobi, (2) Gauss-Lobatto, or (3) Gauss-Radau quadrature. For the first, the *N* abscissas have values inside the interval [0,1], while for the second, the first (ξ_1) and last (ξ_N) abscissas are fixed at 0 and 1, respectively. For the third variant, one abscissa is fixed in mixture-fraction space and could be used, for example, to fix an abscissa at the stoichiometric mixture fraction. For each quadrature rule, weights are fixed such that the mixture-fraction moments agree with those of the corresponding β-PDF. For example, with Gauss-Jacobi quadrature, the number of moments that can be fixed is 2*N*, while, for Gauss-Lobatto quadrature, the number is 2*N*-2. Thus, because we require that at least the first three moments (*j*=0,1,2) of the mixture fraction agree with the β-PDF, the minimum value for *N* is different for each quadrature rule. Namely, with Gauss-Jacobi or Gauss-Radau quadrature, we can use as few as *N*=2 nodes, while at least *N*=3 are required for Gauss-Lobatto quadrature.

The quadrature-based algorithm using SA-CMC for molecular mixing is shown step by step in Fig. 1. Step 1 provides the updated values of the species moments $\langle \xi^k Q \rangle$ and mixture fraction moments $\langle \xi \rangle$ and $\langle \xi^2 \rangle$ at the current time t^n in each grid cell of the computational domain. The remaining steps (2-13) are applied separately for each grid cell and, thus, here, we will describe the algorithm for one cell. In step 2, the β-PDF for the mixture fraction yields the corresponding *N* weights and abscissas. The value of N is arbitrary and is decreased as the solution advances according to the values of the weights such that if the weight on any node is less than 10^{-10}, the Gauss-Lobatto routine in step 2 is run again with $N-1$ nodes. Subsequently, steps 3-13 continue with $N-1$ conditional moments. After computing the quadrature rule in step 2, CQMOM is applied in step 3 to find the conditional means Q_i from the joint moments $\langle \xi^k Q \rangle$. Then, in step 4, the values of the deviation variables are computed, followed by the expansion coefficients A_n in step 5. After the time variable has been updated in step 6, the mixture-fraction moments are updated in step 7 by solving Eq. (2.6). The Gauss-Lobatto quadrature routine is then applied with

the updated moments to find the updated mixture-fraction weights and abscissas in step 8. Steps 9 and 10 compute the updated deviation variable q^* evaluated at the mixture-fraction abscissas, and step 11 converts the deviation variable into the conditional chemical species concentration. After steps 3-5, 10, and 11 have been carried out for each chemical species, a stiff ODE solver is employed to update all of the Q_i due to chemical reactions in step 12. Note that this step is done separately for each quadrature node (i.e., it can be computed in parallel). Finally, in step 13, the updated joint moments are computed from their definition in terms of the quadrature nodes given in Eq. (2.14). Once steps 2-13 have been completed for every grid cell, the algorithm returns to solving the spatial transport equations for the unconditional moments in step 1.

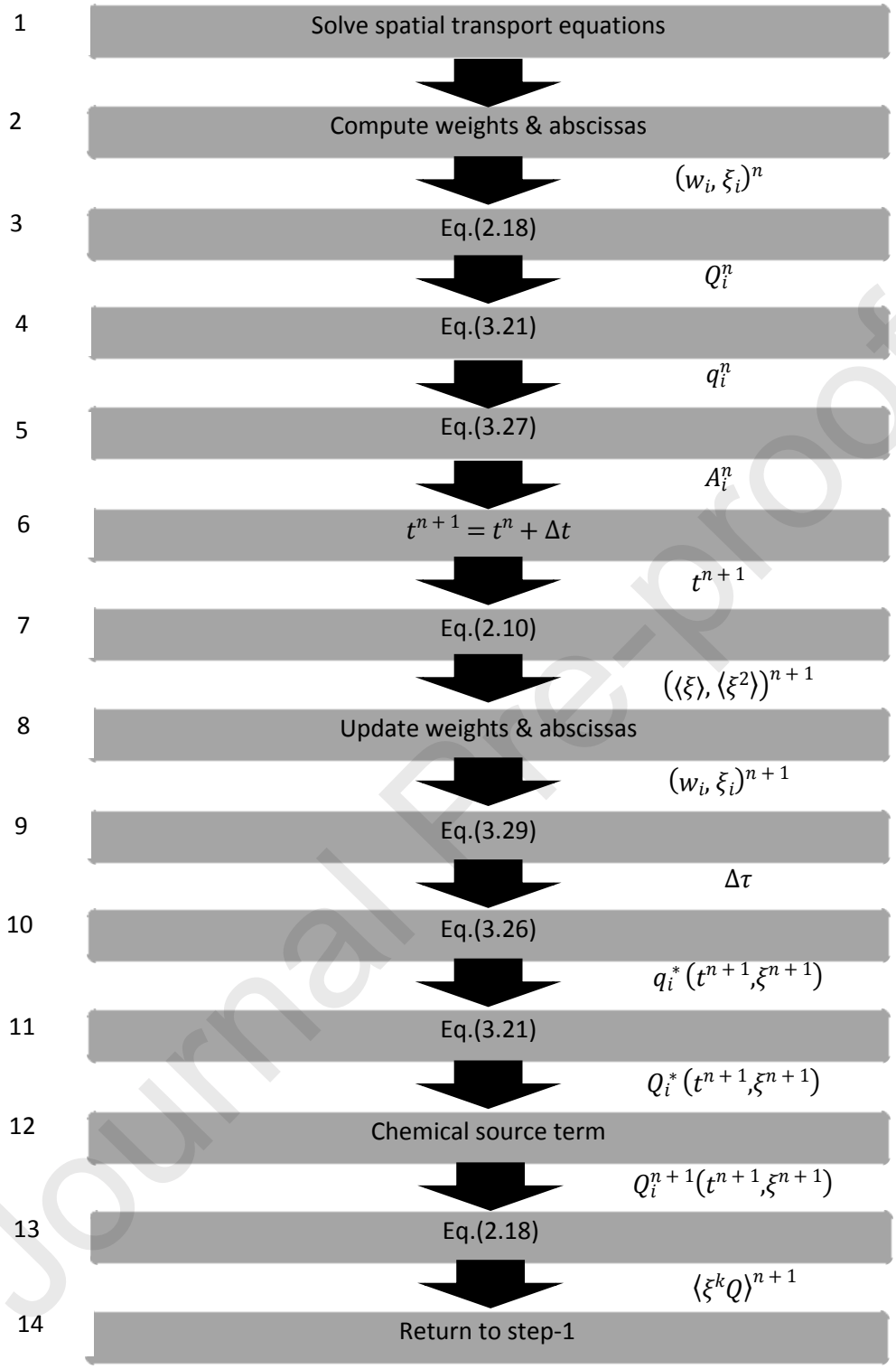


Figure 1 Quadrature-based algorithm for spatial transport, molecular mixing and chemical reactions. In a CFD simulation, the molecular mixing and chemical reaction substeps (2-13) are applied for each grid cell at each time step. The molecular mixing substeps (2-11) are applied for each chemical species, all of which are coupled due to chemical reactions (step 12). The superscript *n* denotes the time step, while * denotes an intermediate value used for the operator spitting.

5. Test Cases with Mixing-Sensitive Reactions

Mixing-sensitive reactions are of great interest, especially in chemical engineering, since in most multi-step reactions, one or more reactions have very fast reaction rates, while the slowest reaction determines the rate of production of undesired byproducts (Bourne 2003). Moreover, the slow reaction is often affected by the mixing efficiency, especially in spatially distributed systems. In this study, steps 2-13 of the quadrature-based algorithm in Fig. 1 have been implemented in MATLAB and tested for two mixing-sensitive reactions: competitive-consecutive and parallel reactions. As is appropriate for liquid-phase, isothermal reactions, the initial conditions for the joint moments were taken to be completely unmixed with two inlet streams.

As described by Fox (2003), for the following competitive-consecutive chemistry

$$A + B \xrightarrow{k_{1,c}} R \tag{3.31}$$

$$B + R \xrightarrow{k_{2,c}} S \tag{3.32}$$

two reaction-progress variables Y_1, Y_2 and the mixture fraction ξ suffice to find the species concentrations. However, in the limit of $k_{1c}^* = k_{1,c} C_{B0} \to \infty$, where $[B]_0$ is the initial concentration of species *B*, the first reaction-progress variable Y_1 can be written in terms of Y_2 and ξ, and the non-negative reaction rate for Y_2 has the form

$$\Im\{Y_{2\infty}\} = \xi_{st} k_{2c}^* h_1(Y_{2\infty}, \xi) h_2(Y_{2\infty}, \xi) \tag{3.33}$$

where $k_{2c}^* = k_{1,c}[B]_0$, $h_1(Y_{2\infty}, \xi) = \dfrac{1-\xi}{1-\xi_{st}} - Y_{2\infty}$ and $h_2(Y_{2\infty}, \xi) = \dfrac{\xi - \xi_{st}}{\xi_{st}(1-\xi_{st})} - Y_{2\infty}$ with $\xi_{st} = [A]_0 / ([A]_0 + [B]_0)$. For values of Y_2 and ξ for which Eq. (3.14) is negative, the reaction rate is zero.

Similarly, for the parallel reaction scheme

$$A + B \xrightarrow{k_{1,p}} R \tag{3.34}$$

$$A + C \xrightarrow{k_{2,p}} S \tag{3.35}$$

with $k_{1p}^{*} = k_{1,p}\left[B\right]_0 \to \infty$, the first reaction-progress variable $Y_{1,p}$ can again be written in terms of $Y_{2,p}$ and ξ, and its reaction rate has the form (Fox 2003)

$$\Im\left\{Y_{2\infty}\right\} = \xi_{s2} k_{2p}^{*} h_1\left(Y_{2\infty},\xi\right) h_2\left(Y_{2\infty},\xi\right) \tag{3.36}$$

where $k_{2p}^{*} = k_{2,p}\left[C\right]_0$ $h_1\left(Y_{2\infty},\xi\right) = \dfrac{\xi}{\xi_{s2}} - Y_{2\infty}$ and $h_2\left(Y_{2\infty},\xi\right) = \dfrac{\xi_{s1}-\xi}{\xi_{s1}\left(1-\xi_{s2}\right)} - Y_{2\infty}$, with $\xi_{s1} = \dfrac{\left[A\right]_0}{\left[A\right]_0+\left[B\right]_0}$ and $\xi_{s2} = \dfrac{\left[A\right]_0}{\left[C\right]_0+\left[A\right]_0}$.

6. Results and Discussion

6.1 Verification of SA-CMC

As a preliminary verification exercise, the SA-CMC algorithm with Gauss-Lobatto quadrature is tested without chemical reactions (pure mixing) against the numerical solution to the Jacobi differential equation (Eq. 3.3) found using the 1-D partial differential equation (PDE) solver (*pdepe*) in MATLAB with two different initial conditions, and boundary conditions $q\left(0,t\right) = q\left(1,t\right) = 0$. Here, the purpose is to test SA-CMC with different initial conditions and with different numbers of nodes for a fixed mixture-fraction mean of $\left\langle\xi\right\rangle = 0.7$. Unless noted otherwise, the initial number of Gauss-Lobatto nodes for SA-CMC was set to 10, while accurate results were obtained with 50 grid points for the PDE solver.

For the first test shown in Figs. 2a and 2b, the initial condition is $q\left(\zeta,0\right) = \left(1-\zeta\right)^{r-1} - \left(1-\zeta\right)^{r} / q_{\max}$ where $q_{\max} = \xi_{\max}^{r-1} - \xi_{\max}^{r}$, $\xi_{\max} = 1-1/r$ and $r = 20$. Note that while this initial condition is `artificial' in the sense that *q* was not produced by a chemical reaction, it does allow us to check that the algorithm is working properly (i.e. conserves the mean value and abscissas are realizable). The first and last time steps are plotted in Fig. 2a, and the time variation of deviation moments $\left\langle\xi^k q\right\rangle$ with *k*=0,1,2,3 are compared for the two solution methods in Fig. 2b. As can be seen from these plots, both methods give similar results, but the decrease in the deviation moments is sharper at the beginning of the simulation with the PDE solver. This difference is mainly because the SA-CMC solution assumes that the mixture fraction depends on time only

through its shape parameters, which is true when the rate of change of σ is small. At the beginning of the simulation, σ is very large, so that the corresponding change in the shape parameters (*a* and *b*) is relatively rapid. However, as can be seen in Fig. 2b, after the first few timesteps, this difference does not result in a significant error, and the deviation moments from both methods reach the same value after a short time.

In Fig. 2c three different numbers of nodes (8, 10, and 20) were tested for the initial condition $q(\zeta,0)=\zeta^{r-1}-\zeta^{r}/q_{\max}$. From Figs. 2c, it can be observed that at most 8 nodes are sufficient when SA-CMC is used, even at the initial times. Furthermore, during the SA-CMC solution, the number of nodes decreased with time (see discussion in Sec. 4) when approaching well-mixed conditions. As expected, the abscissas move towards the constant mean mixture fraction, as shown in Fig. 2d. This also allows computational savings as only a few nodes are required when the abscissas are close to each other (i.e., in the case of a well-mixed reactor). Well mixedness is attained when the mixture-fraction variance decreases to values below 2.5×10^{-4}. As expected, the final quadrature rule is very close to a Dirac delta function centered at $\langle\xi\rangle$, which shows that the SA-CMC algorithm for molecular mixing is accurate even with just a few nodes. Finally, it can be observed in Figs. 2a and 2c that the conditional abscissas Q_i are always positive. This observation confirms that truncating the Jacobi polynomial expansion to find the SA-CMC solution does not result in uncontrollable oscillations or any other undesirable behavior. Nonetheless, at short times the larger value of *N* is required to avoid negative Q_i values. In order to verify that the favorable properties of SA-CMC seen with pure mixing carry over to reacting cases, we next apply SA-CMC with a chemical source term by using operator splitting.

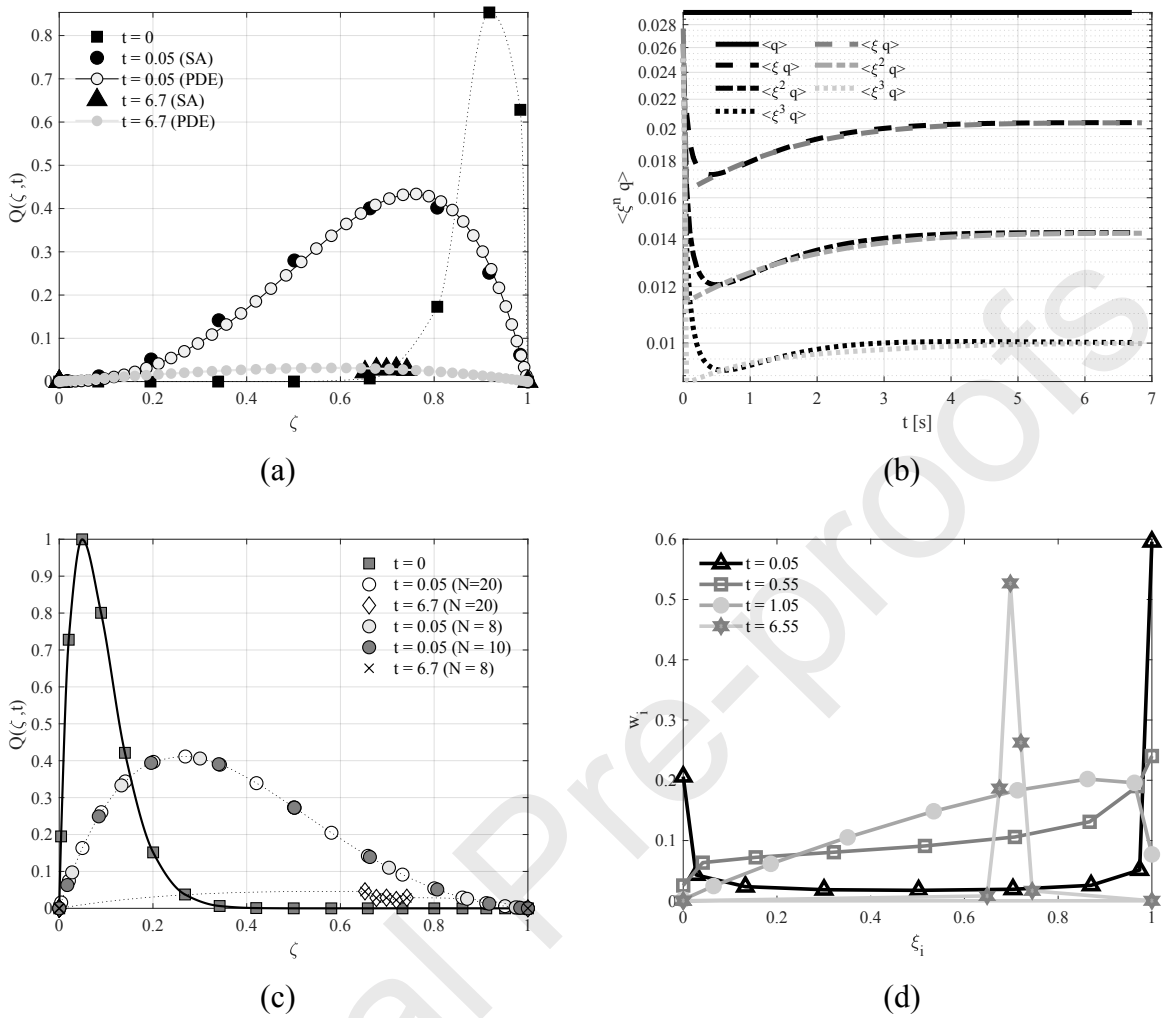


Figure 2. Test cases for pure mixing with mean mixture fraction of 0.7. (a) Conditional means at t = 0, 0.05 and 6.7 s using SA-CMC (*N*=10) and the PDE solver. (b) First four deviation moments. (c) Comparisons of SA-CMC starting with different number of nodes. (d) Gauss-Lobatto quadrature points vs. weights at the corresponding times in (c) with the points connected by straight lines to aid comparison.

6.2 Application of quadrature-based SA-CMC to mixing sensitive reactions

In this section, two mixing-sensitive reaction schemes: competitive-consecutive (CC) and parallel (P) reactions are studied. As explained in Sec. 5, the first reaction-progress variable is written in terms of the second reaction-progress variable Y_2 and mixture fraction. For the chemical source term, Eqs. (3.33) or (3.36) are employed for the CC and P cases, respectively. In the plot representing the allowable values of mixture fraction and progress variable shown in Fig. 3a, the chemical source term is non-zero inside the triangular region represented by the solid lines and zero elsewhere, and the dashed line is the mixing line indicating the upper limit of the progress variable (Fox 2003). Thus, in order to be physically valid, the SA-CMC solution for the conditional

moment of Y_2 must remain between the ξ axis and the upper limit given by the dashed/solid lines at all times. Note that, from the initial conditions, Y_2 is zero at time zero and will increase only due to the chemical source term.

The triangular region that bounds the allowable region is defined analytically in terms of the maximum achievable reaction-progress variable $Y_{2\max}(Q_{\max})$ (given by the gray circle on the y axis) and the corresponding mixture fraction $\xi_{\max}$ (given by the gray circle on the x axis). For the CC case, these points are found from

$$\xi_{\max} = \frac{2\xi_{st}}{1+\xi_{st}}, \qquad Y_{2\max} = \frac{1}{1+\xi_{st}} = \frac{1}{2}\frac{\xi_{\max}}{\xi_{st}} \tag{4.38}$$

with $\xi_{st} = [A]_0 / \left([B]_0 + [A]_0\right)$. For P case, they are defined as

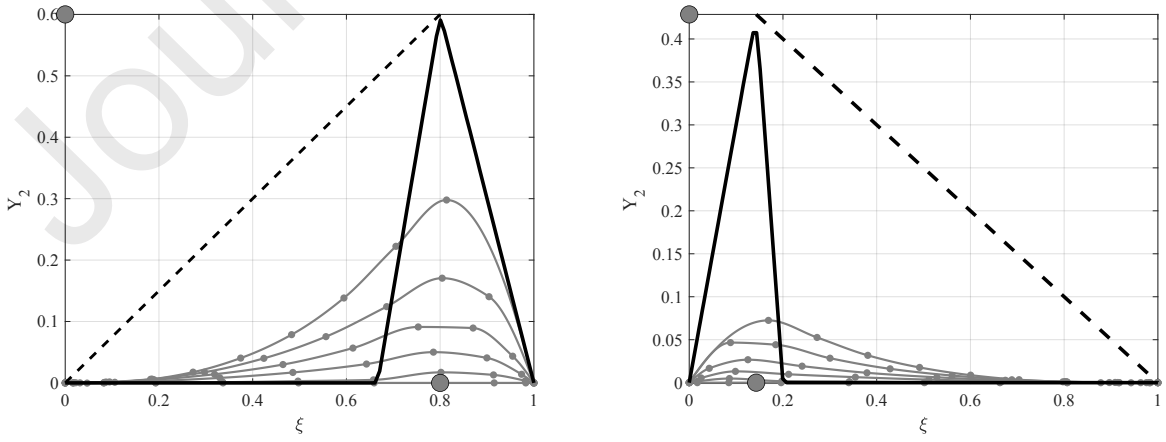
$$\xi_{\max} = \frac{\xi_{s1}\xi_{s2}}{\xi_{s2} + \xi_{s1}(1-\xi_{s2})} = \frac{[A]_0}{[A]_0 + [B]_0 + [C]_0}, \quad Y_{2\max} = \frac{\xi_{s1}}{\xi_{s2} + \xi_{s1}(1-\xi_{s2})} = \frac{\xi_{\max}}{\xi_{s2}} \tag{4.39}$$

where $\xi_{s1} = [A]_0 / \left([A]_0 + [B]_0\right)$ and $\xi_{s2} = [A]_0 / \left([C]_0 + [A]_0\right)$. In our study, the initial concentrations are selected to be $[A]_0 = 2$, $[B]_0 = 1$ and $\langle\xi\rangle = 0.25$ for the CC case, and $[A]_0 = 0.5$ $[B]_0 = 2$, $[C]_0 = 1$ and $\langle\xi\rangle = 0.75$ for the P case. The corresponding stoichiometric mixture fractions are $\xi_{st} = 0.67$ and $\xi_{s1} = 0.2$, $\xi_{s2} = 0.33$. From Eqs. (4.38) and (4.39) the triangular regions are then defined by $\xi_{max} = 0.80$, $Y_{2max} = 0.6$ and $\xi_{max} = 0.143$, $Y_{2max} = 0.43$ for the CC and P cases, respectively. The Damköhler number for the finite-rate chemical reaction is defined by $Da = \tau_\xi / \tau_\alpha$ where $\tau_\alpha = 1/k_2^*$ is the reaction time scale. Unless indicated otherwise, $Da = 2$ in the simulations below.

The initial concentrations above were selected such that, if the mixing were perfect (i.e., no segregation due to mixing), a limited amount of *B* is present in the CC case, and thus all *B* would be consumed instantaneously via the first reaction, in which case no byproduct *S* would be produced. On the other hand, when mixing is not instantaneous relative to the reaction rate and results in segregation, the primary product *R* will react with *B* to produce *S*. In other words, the

amount of *S* produced is an indication of the degree of segregation. Thus, for reactive mixing, the production of *S* is the first criterion for testing SA-CMC. Example results illustrating this behavior for the CC case are given in Figs. 3a, 3c, and 3e. For the P case, similar conclusions can be drawn from Figs. 3b, 3d, and 3f. However, as the species *A* is consumed in both reactions for the P case, the concentration of *A* decreases rapidly, as is seen in Fig. 3d. For this reason, *A* is the limiting species, and if mixing is not perfect, a local excess of *A* will produce *S*. Therefore, as in the CC case, the amount of *S* is a measure of the segregation intensity.

An equally important criterion for the SA-CMC solution is that the conditional moments of the reaction-progress variable (given by the gray lines in Figs. 3a and 3b) do not exceed the upper limit defined by the reaction/mixing line. In Fig. 3, we can see that all of the conditional means are within the triangular region and approach the mixing line as the reaction proceeds. Furthermore, as can be seen from Fig. 3c, a relatively small amount of *S* is produced, and the reactions end when *B* is completely consumed. Hence, as expected for this Damköhler number, the mixing is not "perfect", and species *S* is mainly produced within the first 1.5 s. It also can be seen that after approximately 1.5 s the reaction rate is low, which can also be observed from the deviation moments for the reaction-progress variable in Fig. 3e. By definition, $\langle q \rangle$ changes only due to the chemical reaction, while its value is constant in the absence of the chemical reaction. From Fig. 3e it is observed that in the initial 1.5 s, the value of $\langle q \rangle$ increases and then stays nearly constant as mixing continues. However, the higher-order moments continue to change for larger times due to mixing.



(a) (b)

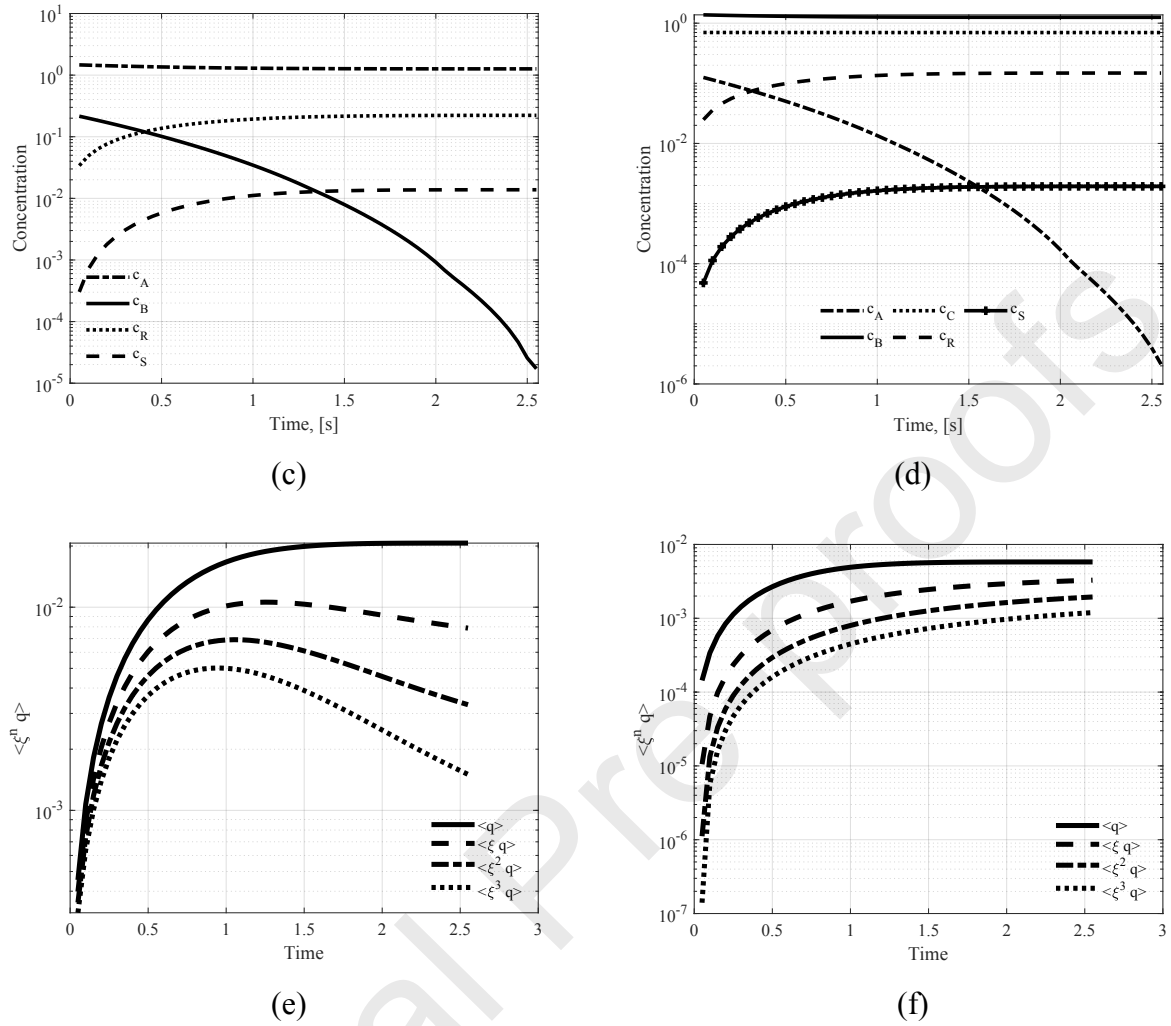
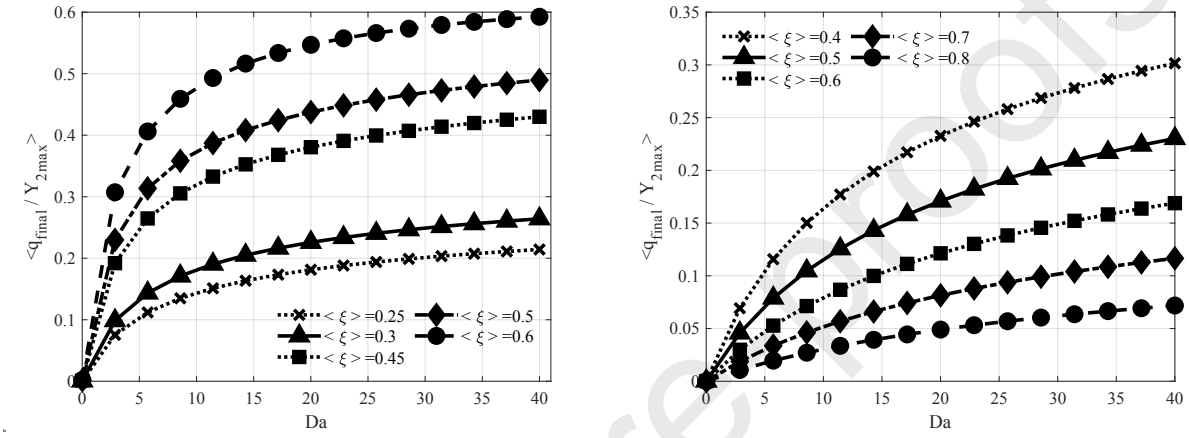


Figure 3. Results for competitive-consecutive (left column) and parallel reactions (right column) with N=10. (a-b) Conditional scalar means at increasing times where abscissas are shown on the corresponding curve. (c-d) Concentrations of chemical species vs. time. (e-f) Time evolution of the first four deviation moments of S.

After confirming that the quadrature-based algorithm behaves as expected for the selected chemical time scale and mixture fraction mean, it was tested over a range of chemical timescales with different values of $\langle \xi \rangle$ for the CC and P cases. The Damköhler number range was selected to cover values representing rapid mixing up to the reaction-controlling limit, i.e., 15 values of Da linearly spaced between 0.005 and 40 were tested. Five different mean mixture fractions were investigated: $\langle \xi \rangle = [0.25, 0.3, 0.45, 0.5, 0.6]$ for CC and $\langle \xi \rangle = [0.4, 0.5, 0.6, 0.7, 0.8]$ for P, respectively. The results for all of the different cases are summarized in Fig. 4. As expected, for CC (Fig. 4a), the ratio of $Y_2 / Y_{2\max}$ exponentially increases to reach its corresponding limiting value

with increasing *Da*, and for the largest $\langle\xi\rangle$ the limiting value is the highest. Likewise, for P (Fig. 4b), the opposite dependence on $\langle\xi\rangle$ is observed, as is expected from the location of the non-zero reaction rate in Fig. 3b. In other words, for a given Damköhler number, a larger amount of undesired product will be produced when the mean mixture fraction is nearer to the non-zero reaction zone.



(a) (b)

Figure 4. Final values of *Q/Qmax* versus Damköhler number (*Da*) for different mean mixture fractions with *N*=10. (a) Competitive-consecutive reactions. (b) Parallel reactions. The value of *Q/Qmax* is proportional to the fraction of the fluid experiencing mixing segregation during the reactions.

7. Conclusions

In CMC applications to simulate turbulent reacting flows using CFD, two separate grids must be generated, i.e., in 3-D physical space and for the 1-D conditioning-variable space. In practice, to conserve the scalar mean during molecular mixing, a relatively fine conditioning-scalar grid must be employed, which drastically increases the computational cost relative to CFD simulations employing moment closures. In this study, the requirement for the additional grid in mixture-fraction space is eliminated by proposing a solution algorithm for molecular mixing that uses a quadrature-based, semi-analytical solution expressed in terms of Jacobi polynomials. The conditioning on mixture fraction is represented by the joint mixture-fraction, chemical-concentration moments, and is reconstructed from these moments via Gaussian quadrature rules. The semi-analytical solution is defined such that the unconditional mean $\langle q \rangle$ is constant during molecular mixing, regardless of the number of quadrature nodes. As with the ME-PDF approach, the quadrature nodes offer a convenient representation of the chemical composition of the fluid

from which it is straightforward to evaluate the changes due to chemical reactions of arbitrary complexity. Indeed, the quadrature-based algorithm with the Gauss-Jacobi quadrature can be viewed as the logical extension of ME-PDF methods to more than two environments.

As the SA-CMC algorithm treats the molecular mixing part of the CMC equation, the quadrature-based algorithm was first verified for pure mixing by comparing its solution to the numerical solution of the Jacobi differential equation (Eq. (3.22)). The quadrature-based algorithm was then tested with chemical source terms for the mixing-sensitive, competitive-consecutive and parallel reaction schemes. In every test case investigated, physically relevant results were obtained for the conditional moments. In addition, it was found that at most ten quadrature nodes are required to accurately represent the conditional moments, with the possibility of decreasing the number of nodes as the well-micromixed condition is approached. By applying the quadrature-based algorithm over a wide range of Damköhler numbers and mixture-fraction means, it was demonstrated that SA-CMC treatment of molecular mixing is a robust alternative to grid-based methods for first-order CMC applications. Future work should therefore focus on applying the quadrature-based algorithm to more complex reaction schemes such as turbulent combustion, and on coupling it to a CFD code to simulate spatially inhomogeneous turbulent reacting flows. In addition, for cases where the conditioning variable cannot be well represented by a β-PDF (e.g., multiphase flows such as spray combustion), the CQMOM methodology introduced in this work can be extended to arbitrary forms for the mixture-fraction PDF by solving additional transport equations for the mixture-fraction moments.

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HIGHLIGHTS

- A quadrature-based semi-analytical solution algorithm (SA-CMC) for first-order conditional moment closure (CMC) applications is proposed and tested.
- SA-CMC eliminates the need for an additional grid in mixture-fraction space in CMC applications.
- SA-CMC is first verified for pure mixing, and then applied to mixing-sensitive reactions.
- SA-CMC is an extension of multi-environment probability density function (ME-PDF) method to more than two environments.

CRediT authorship contribution statement

Aziz D. Ilgun: Verification of the algorithm, implementation of the simulations for the test cases, writing the first draft of the paper. **Alberto Passalacqua:** Advising, proofreading and editing on the final version of the paper. **Rodney O. Fox:** Original idea, testing of the first version of the algorithm; advising and troubleshooting on the final version of the algorithm; writing and editing of the paper.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.